

### Claim Rejections – 35 USC 103

#### *Sugimura, et al*

Sugimura discloses a low temperature catalyst step at column 53, lines 13-20 and also at column 54, lines 51-59. These steps comprise a compound (b) that is a compound of a transition metal from any of Groups 8 to 10 of the periodic table. See column 47, lines 25-37.

- The compounds described in Sugimura, a transition metal from Groups 8 to 10 of the periodic table, would not be considered a metallocene catalyst as required in the present invention.

Sugimura does not disclose including compound (e), a "fine particulate carrier", in the low temperature catalyst step.

- The present invention requires "a particulate catalyst support material comprising silica particles impregnated with an alumoxane co-catalyst ..." in step (a) of the synthesis.

Sugimura does not teach the percentage of impregnation of the internal pore volume of silica.

#### Double Patenting

The double patenting rejections relating to U.S. Patent No.'s 6,239,058, 6,166,153, 5,968,864, and 6,432,860 are overcome by the following differences:

- 6,239,058      The catalyst of this patent is dried, as disclosed at column 5, line 10 and claim 1, column 12, line 12. It does not teach the benefits of low temperature synthesis and of the benefits of elimination of the drying step. It also does not teach the percentage of impregnation of the internal pore volume of the support.
- 6,166,153      This patent does not teach the benefits of low temperature synthesis and of the benefits of elimination of the drying step. It also does not teach the percentage of impregnation of the internal pore volume of the support.
- 5,968,864      The catalyst of this patent can be dried, as disclosed at column 5, line 15 and claim 1, column 11, line 6. This patent does teach the benefits of low

temperature synthesis, but does not teach the benefits of elimination of the drying step. It also does not teach the percentage of impregnation of the internal pore volume of the support.

- 6,432,860 This patent does not teach the benefits of low temperature synthesis and of the benefits of elimination of the drying step.

## CLAIMS

1. (original) A process for the preparation of a supported metallocene catalyst comprising:

(a) providing a particulate catalyst support material comprising silica particles impregnated with an alumoxane co-catalyst with at least one-half of said co-catalyst disposed within the internal pore volume of said silica particles;

*A1* (b) providing a dispersion in an aromatic hydrocarbon solvent of a stereospecific metallocene incorporating a metallocene ligand structure having two sterically dissimilar cyclopentadienyl ring structures coordinated with a central transition metal atom; at least one of said cyclopentadienyl ring structures being a substituted cyclopentadienyl group which provides an orientation with respect to said transition metal atom which is sterically different from the orientation of the other cyclopentadienyl group with respect to said transition metal atom, and both of said cyclopentadienyl groups being in a relationship with each other providing a stereorigid relationship relative to said coordinating transition metal atom to prevent rotation of said ring structures;

(c) mixing said metallocene-solvent dispersion and said alumoxane-impregnated silica particles at a temperature of about 10°C or less for a period sufficient to enable said metallocene to become reactively supported on and impregnated within said alumoxane- impregnated silica particles and form a silica-supported catalyst;

(d) recovering said supported catalyst from said aromatic solvent;

(e) washing said supported catalyst with a paraffinic hydrocarbon solvent at a temperature of about 10°C or less; and

(f) dispersing said washed catalyst in a viscous mineral oil having a viscosity greater than the viscosity of said paraffinic hydrocarbon solvent.

2. (original) The process of claim 1 wherein the washed catalyst at the time of dispersion in said viscous mineral oil contains a residual amount of said paraffinic hydrocarbon solvent.

*Q1 Cont*  
3. (original) The process of claim 2 wherein the washing of said supported catalyst with said paraffinic hydrocarbon solvent is effective to reduce the residual aromatic solvent on said supported catalyst to a value of no more than 50 wt.% of said supported catalyst.

4. (original) The process of claim 1 wherein said viscous mineral oil has a viscosity of at least 10 centistokes.

5. (currently amended) The ~~method~~ process of claim 4 wherein said paraffinic hydrocarbon solvent has a viscosity of no more than 2 centistokes.

6. (original) The process of claim 1 wherein subsequent to step (d) and prior to step (e) washing the recovered supported metallocene catalyst with an aromatic solvent to remove unsupported metallocene from said supported metallocene catalyst and thereafter washing said supported catalyst in accordance with step (e).

7. (original) The process of claim 1 wherein said silica support material comprises spheroidal silica particles having an average particle size within the range of 20-60 microns.

8. (original) The process of claim 7 wherein said spheroidal silica particles have an average effective pore diameter within the range of 200-400 Angstroms.

9. (original) The process of claim 8 wherein the weight ratio of said alumoxane to said silica in subparagraph (a) of claim 1 is within the range of about 0.6-2.0.

10. (original) The process of claim 1 wherein said stereospecific metallocene is a syndiospecific metallocene characterized by the formula:



wherein  $Cp_a$  is a substituted cyclopentadienyl ring,  $Cp_b$  is an unsubstituted or substituted cyclopentadienyl ring; each R is the same or different and is a hydrocarbyl radical having 1-20 carbon atoms; each  $R'_m$  is the same or different and is a hydrocarbyl radical having 1-20 carbon atoms;  $R''$  is a structural bridge between the cyclopentadienyl rings imparting stereorigidity to the metallocene and is selected from the group consisting of an alkylene radical having 1-4 carbon atoms, a silicon hydrocarbyl radical, a germanium hydrocarbyl radical, a phosphorus hydrocarbyl radical, a nitrogen hydrocarbyl radical, a boron hydrocarbyl radical, and an aluminum hydrocarbyl radical; Me is a group 4b, 5b, or 6b metal from the Periodic Table of Elements; each Q is a hydrocarbyl radical having 1-20

carbon atoms or is a halogen;  $0 \leq p \leq 3$ ;  $0 \leq m \leq 4$ ;  $1 \leq n \leq 4$ ; and wherein  $R'_m$  is selected such that  $(Cp_bR'_m)$  is a sterically different ring than  $(Cp_aR_n)$ .

11. (original) The process of claim 10 wherein R is selected such that  $(Cp_aR_n)$  forms a substituted or unsubstituted fluorenyl group.

12. (original) The process of claim 11 wherein Me is titanium, zirconium, hafnium, or vanadium.

Al  
Cont

13. (original) The process of claim 12 wherein  $R''$  is a methylene, ethylene, organosilyl, substituted methylene, or substituted ethylene radical.

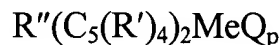
14. (original) The process of claim 13 wherein R is selected such that  $(Cp_aR_n)$  forms a substituted or unsubstituted fluorenyl radical having bilateral symmetry and  $R'$  is selected such that  $(Cp_bR'_m)$  forms an alkyl substituted or unsubstituted cyclopentadienyl radical having bilateral symmetry.

15. (original) The process of claim 14 wherein  $R''(Cp_aR_n)(Cp_bR'_m)$  forms an isopropylidene(cyclopentadienyl-1-2,7-di-tert-butyl fluorenyl) ligand or a diphenyl methylene(cyclopentadienyl-1-fluorenyl) ligand.

16. (original) The process of claim 10 wherein said stereospecific metallocene is selected from the group consisting of isopropylidene(cyclopentadienyl-1-2,7-

di-tert-butyl fluorenyl) zirconium dichloride and diphenyl methylene(cyclopentadienyl-1-fluorenyl) zirconium dichloride and mixtures thereof.

17. (original) The process of claim 1 wherein said stereospecific metallocene is an isospecific metallocene characterized by the formula:



*Al Cont*  
wherein each  $(C_5(R')_4)$  is a substituted cyclopentadienyl ring; each  $R'$  is the same or different and is a hydrogen or hydrocarbyl radical having 1-20 carbon atoms;  $R''$  is a structural bridge between the two  $(C_5(R')_4)$  rings imparting stereorigidity to said metallocene with the two  $(C_5(R')_4)$  rings being in a racemic configuration relative to Me, and  $R''$  is selected from the group consisting of an alkylene radical having 1-4 carbon atoms, a silicon hydrocarbyl radical, a germanium hydrocarbyl radical, a phosphorus hydrocarbyl radical, a nitrogen hydrocarbyl radical, a boron hydrocarbyl radical, and an aluminum hydrocarbyl radical; Me is a group 4b, 5b, or 6b metal as designated in the Periodic Table of Elements; each Q is a hydrocarbyl radical having 1-20 carbon atoms or is a halogen; and  $0 \leq p \leq 3$ .

18. (currently amended) The ~~method~~ process of claim 17 wherein said  $(C_5(R')_4)$  groups are indenyl groups which are substituted or unsubstituted.

19. (currently amended) The ~~method~~ process of claim 18 wherein said indenyl groups are each substituted at the proximal position.

20. (currently amended) The ~~method~~ process of claim 18 wherein Me is titanium, zirconium, hafnium, or vanadium.

21. (currently amended) The ~~method~~ process of claim 20 wherein R" is a methylene, ethylene, organosilyl, substituted methylene, or substituted ethylene radical.

22. (currently amended) The ~~method~~ process of claim 21 wherein said isospecific metallocene is selected from the group consisting of racemic dimethylsilyl(2-methyl-4-phenyl indenyl)<sub>2</sub> zirconium dichloride, racemic dimethylsilyl(2-methyl-4-indenyl)<sub>2</sub> zirconium dichloride, and racemic dimethylsilyl(2-methyl-4,5-benzo indenyl)<sub>2</sub> zirconium dichloride and admixture thereof.

23. (original) A process for the preparation of a supported metallocene catalyst comprising:

(a) providing a particulate catalyst support material comprising spheroidal silica particles having an average particle size within the range of 20-60 microns and an average effective pore diameter within the range of 200-400 Angstroms;

(b) contacting said particulate support material with an alumoxane co-catalyst in an aromatic carrier liquid;

(c) heating said mixture of support, carrier liquid, and alumoxane co-catalyst at an elevated temperature for a period sufficient to fix said alumoxane on said particulate support material with at least one-half of said co-catalyst disposed within the internal pore volume of said silica particles;

(d) cooling said mixture and separating said alumoxane-containing support material from said carrier liquid;

(e) washing said alumoxane containing support material with an aromatic solvent to remove excess alumoxane therefrom;



Al Cont (f) cooling said alumoxane containing support material to a reduced temperature of about 10°C or less and at said reduced temperature adding to said support material a dispersion in an aromatic hydrocarbon solvent of a stereospecific metallocene incorporating a metallocene ligand structure having two sterically dissimilar cyclopentadienyl ring structures coordinated with a central transition metal atom; at least one of said cyclopentadienyl ring structures being a substituted cyclopentadienyl group which provides an orientation with respect to said transition metal atom which is sterically different from the orientation of the other cyclopentadienyl group with respect to said transition metal atom, and both of said cyclopentadienyl groups being in a relationship with each other providing a stereorigid relationship relative to said coordinating transition metal atom to prevent rotation of said ring structures;

(g) mixing said metallocene-solvent dispersion and said alumoxane-impregnated silica particles at a temperature of about 10°C or less for a period sufficient to enable said metallocene to become reactively supported on and impregnated within said alumoxane- impregnated silica particles and form a silica-supported catalyst;

(h) recovering the resulting supported metallocene catalyst from said aromatic solvent;

(i) washing said supported catalyst with a paraffinic hydrocarbon solvent at a reduced temperature of about 10°C or less; and

(j) thereafter dispersing said supported metallocene in a viscous mineral oil

having a viscosity substantially greater than the viscosity of said paraffinic hydrocarbon solvent.

24. (original) The process of claim 23 wherein subsequent to step (h) and prior to step (i) washing the recovered supported metallocene catalyst with an aromatic solvent to remove unsupported metallocene from said supported metallocene catalyst and thereafter washing said supported catalyst in accordance with step (i).

25. (original) The process of claim 23 wherein the weight ratio of said alumoxane to said silica is within the range of about 0.6-2.0.

26. (original) The process of claim 23 wherein said stereospecific metallocene is a syndiospecific metallocene characterized by the formula:

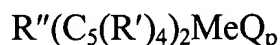


wherein  $Cp_a$  is a substituted cyclopentadienyl ring,  $Cp_b$  is an unsubstituted or substituted cyclopentadienyl ring; each  $R$  is the same or different and is a hydrocarbyl radical having 1-20 carbon atoms; each  $R'_m$  is the same or different and is a hydrocarbyl radical having 1-20 carbon atoms;  $R''$  is a structural bridge between the cyclopentadienyl rings imparting stereorigidity to the catalyst and is selected from the group consisting of an alkylene radical having 1-4 carbon atoms, a silicon hydrocarbyl radical, a germanium hydrocarbyl radical, a phosphorus hydrocarbyl radical, a nitrogen hydrocarbyl radical, a boron hydrocarbyl radical, and an aluminum hydrocarbyl radical;  $Me$  is a group 4b, 5b, or 6b metal from the Periodic Table of Elements; each  $Q$  is a hydrocarbyl radical having 1-20 carbon atoms or is a halogen;  $0 \leq p \leq 3$ ;  $0 \leq m \leq 4$ ;  $1 \leq n \leq 4$ ; and wherein  $R'_m$  is selected such that  $(Cp_bR'_m)$  is a sterically different ring than  $(Cp_aR_n)$ .

27. (original) The process of claim 26 wherein the weight ratio of said alumoxane to said silica is within the range of about 0.6-2.0.

28. (original) The process of claim 27 wherein the concentration of said metallocene on said support is within the range of 0.1-6 wt.%.

29. (currently amended) The ~~method~~ process of claim 23 wherein said stereospecific metallocene is a chiral stereorigid metallocene characterized by the formula:



Al  
Cord  
wherein each  $(C_5(R')_4)$  is a substituted cyclopentadienyl ring; each  $R'$  is the same or different and is a hydrogen or hydrocarbyl radical having 1-20 carbon atoms;  $R''$  is a structural bridge between the two  $(C_5(R')_4)$  rings imparting stereoridity to said catalyst with the two  $(C_5(R')_4)$  rings being in a racemic configuration relative to Me, and  $R''$  is selected from the group consisting of an alkylene radical having 1-4 carbon atoms, a silicon hydrocarbyl radical, a germanium hydrocarbyl radical, a phosphorus hydrocarbyl radical, a nitrogen hydrocarbyl radical, a boron hydrocarbyl radical, and an aluminum hydrocarbyl radical; Me is a group 4b, 5b, or 6b metal as designated in the Periodic Table of Elements; each Q is a hydrocarbyl radical having 1-20 carbon atoms or is a halogen; and  $0 \leq p \leq 3$ .

30. (original) The process of claim 29 wherein the weight ratio of said alumoxane to said particulate silica support is within the range of about 0.6-2.0.

31. (original) The process of claim 30 wherein the concentration of said metallocene on said support is within the range of 0.1-6.0 wt.%.

---

Applicants respectfully submit the enclosed amendments and believe that the pending claims as amended are now in condition for allowance.

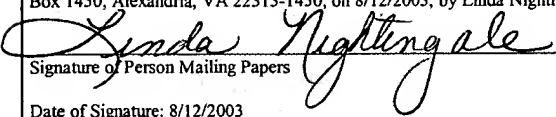
Respectfully submitted,



Bradley A. Misley  
Reg. No. 46,937  
Fina Technology, Inc.  
PO Box 674412  
Houston, TX 77267-4412  
(281) 227-5370

Mailing Certificate

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE U.S. POSTAL SERVICE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO: COMMISSIONER FOR PATENTS, PO Box 1450, Alexandria, VA 22313-1450, on 8/12/2003, by Linda Nightingale.



Signature of Person Mailing Papers

Date of Signature: 8/12/2003